



AFRL-RW-EG-TP-2013-011

Emission Spectroscopy of the Interior of Optically Dense Post-Detonation Fireballs

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March 2013

Interim Report

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October 26, 2012**

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REPORT DOCUMENTATION PAGE				Form Approved OMB No. 0704-0188	
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1. REPORT DATE (DD-MM-YYYY) 18 March 2013		2. REPORT TYPE Interim		3. DATES COVERED (From - To) May 2012 – September 2012	
4. TITLE AND SUBTITLE Emission Spectroscopy of the Interior of Optically Dense Post-Detonation Fireballs				5a. CONTRACT NUMBER	
				5b. GRANT NUMBER	
				5c. PROGRAM ELEMENT NUMBER 62102F	
6. AUTHOR(S) W.K. Lewis ¹ , C.G. Rumchik ² , M.J. Smith ³				5d. PROJECT NUMBER 4347	
				5e. TASK NUMBER 95	
				5f. WORK UNIT NUMBER 02	
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) ¹ University of Dayton Research Institute, Dayton, OH 45469 ² Air Force Research Laboratory, AFRL/RWME, Eglin AFB, FL 32542 ³ Air Force Research Laboratory, AFRL/RQ, Wright-Patterson AFB, OH 45433				8. PERFORMING ORGANIZATION REPORT NUMBER AFRL-RW-EG-TP-2013-011	
9. SPONSORING / MONITORING AGENCY NAME(S) AND ADDRESS(ES) Air Force Research Laboratory, Munitions Directorate Ordnance Division Energetic Materials Branch (AFRL/RWME) Eglin AFB FL 32542-5910 Branch Technical Advisor: Dr. Michael Lindsay				10. SPONSOR/MONITOR'S ACRONYM(S) AFRL-RW-EG	
				11. SPONSOR/MONITOR'S REPORT NUMBER(S) AFRL-RW-EG-TP-2013-011	
12. DISTRIBUTION / AVAILABILITY STATEMENT Distribution A: Approved for public release; distribution unlimited. Approval Confirmation 88ABW/PA # 88ABW-2012-5733, Dated October 26, 2012					
13. SUPPLEMENTARY NOTES SUBJECT TO EXPORT CONTROL LAWS DISTRIBUTION STATEMENT INDICATING AUTHORIZED ACCESS IS ON THE COVER PAGE AND BLOCK 12 OF THIS FORM. DATA RIGHTS RESTRICTIONS AND AVAILABILITY OF THIS REPORT ARE SHOWN ON THE NOTICE AND SIGNATURE PAGE.					
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15. SUBJECT TERMS Nanoenergetics, nano aluminum, fireball temperature, barium nitrate dopant, emission spectroscopy, post detonation diagnostics					
16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT SAR	18. NUMBER OF PAGES 11	19a. NAME OF RESPONSIBLE PERSON Chad G. Rumchik
a. REPORT UNCLASSIFIED	b. ABSTRACT UNCLASSIFIED	c. THIS PAGE UNCLASSIFIED			19b. TELEPHONE NUMBER (include area code) 850-882-7243

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Emission spectroscopy of the interior of optically dense post-detonation fireballs

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(Received 1 November 2012; accepted 12 December 2012; published online 9 January 2013)

In recent years, emission spectroscopy has been applied to the study of post-detonation combustion in explosives, often yielding valuable information on temperatures and chemical dynamics. The post-detonation fireballs that form as under-oxidized detonation products burn in the surrounding air are optically dense and the corresponding emission spectra sample only the material at or near the surface of the fireball. In the present study, we exploit the large optical density in order to probe the dynamics occurring in the interior of the fireball. Emission spectra are collected following detonation of 20 g aluminized Hexahydro-1,3,5-trinitro-1,3,5-triazine (RDX) charges using fiber optics located behind the flame front and then compared with the corresponding spectra of the surface layer collected from outside the fireball. We find that in the early evolution of the fireball ($t \leq 60 \mu\text{s}$ and $r \leq 10 \text{ cm}$ in the current study), combustion and light emission are predominantly confined to the surface, while the interior is dark. Later, after the fireball expands and mixes with the surrounding air ($t \geq 120 \mu\text{s}$ or $r \geq 30 \text{ cm}$), combustion and emission occur throughout, and we find no significant differences between the spectra collected from the interior of the fireball versus those from its surface. © 2013 American Institute of Physics. [<http://dx.doi.org/10.1063/1.4774029>]

I. INTRODUCTION

Understanding energy release processes and their associated kinetics is of fundamental importance for the study of explosives. A comprehensive understanding of the energy release requires knowledge not only of the time-dependent pressure and temperature in the system but also of the chemical dynamics in the evolving system. Identifying important chemical species (including transients) and tracking their evolution through time can greatly expand our ability to model and perhaps even control energy release.

Measurements of the energy release process of explosives are made difficult by the fast timescales involved in the reaction. For gram-scale samples, detonation is typically completed within several microseconds. Subsequent afterburning of under-oxidized detonation products can then result in a post-detonation fireball that can persist for several milliseconds.¹ Measurement options are further limited by the high temperatures and pressures present during these events since most sensors rugged enough to survive the explosive cannot provide a response fast enough to follow its dynamics. Spectroscopy methods are considered to be a very attractive approach to the study of these systems. Spectroscopy techniques have been extensively employed to study flames² and plasmas,³ yielding a large body of work with which to compare. Additionally, spectroscopy can be used to study very fast processes, and spectra can be collected at a distance from the explosion. A number of studies utilizing spectroscopy in the visible, near-IR, and IR to study explo-

sions have now been reported in recent years.^{4–12} Depending upon the technique employed, spectroscopy data can be used to obtain temperature measurements, chemical dynamics data, or both.

One important aspect of the spectroscopy of explosions has been that of the opacity of the fireball. The results of several investigations have suggested that fireballs are optically thick for at least some of their evolution.^{13–17} More recently, the opacity has been quantified¹⁸ as a function of space and time for aluminized explosives, with characteristic attenuation depths on the order of millimeters for its early evolution and centimeters for the remainder of its lifetime for which significant luminosity is observed. This work clearly establishes (at least for gram-scale and larger samples) that emission spectra collected from post-detonation combustion correspond to the conditions near the surface of the fireball but raises other important questions regarding what is occurring in the interior of the fireball and how it is different than the surface dynamics. The purpose of the current study is to exploit the large optical density in order to probe the dynamics occurring in the interior of the fireball. Time-resolved emission spectra are collected from fiber optics located behind the flame front and then compared with the corresponding spectra of the surface layer collected from fiber optics positioned outside the fireball.

II. EXPERIMENT

The experiment is depicted schematically in Figure 1. Pressed right-cylindrical charges of 20 g total mass were prepared from a mixture of RDX (73 wt. %), a hydroxyl-terminated polybutadiene (HTPB) binder (6 wt. %), and

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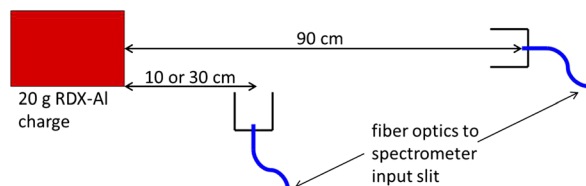


FIG. 1. Schematic representation of the experiment. The cylindrical explosive charges (20 g total mass) consisted of 20 wt. % aluminum nanoparticles in RDX. Light from the expanding fireball was collected by 600 μm core-diameter fiber optics placed relative to the charge as shown (diagram not to scale). The ends of the fibers were enclosed in black cylindrical cups (25 mm width, 25 mm height) in order to limit the field of view.

aluminum nanoparticles (20 wt. %). The aluminum nanoparticles were purchased from Nano Technologies (30–70 nm particle size). In order to obtain temperature measurements, 1 wt. % ball-milled barium nitrate was added to the mixture and mixed thoroughly before pressing. The resulting Ba atomic emission lines can be used to measure the apparent temperature of the flame as demonstrated previously.^{11,12} The charges were detonated from the left side using Reynolds RP-80 detonators placed on the end of each cylindrical charge. Note that in the current investigation, detonation should be complete within 5 μs of detonator initiation given the length of the charge (25 mm) and the detonation velocity of the formulation ($\sim 7.5 \text{ km/s}$), with subsequent emission assigned to the post-detonation fireball resulting from after-burning of under-oxidized detonation products.

Light from the explosions was collected using 600 μm core-diameter fiber optics (Ocean Optics, 38 m total length) as shown in Figure 1. The bare end of the fiber optic was placed inside a black cylindrical cup (25 mm width \times 25 mm height) in order to limit the field of view. The fiber optic was then placed 10, 30, or 90 cm from the right face of the charge. At 10 or 30 cm, the fiber was positioned vertically with no line-of-sight to the charge. At 90 cm, the fiber was placed horizontally to directly view the explosive sample.

Light from the fiber optics was sent to spectrograph located in a shielded observation room several meters away from the explosive charge. The spectrograph was constructed from a 1/8 m spectrometer (Oriel) interfaced to a 4096 pixel line-scan camera (Basler Sprint) with a data collection rate of 1–70 kHz. The resolution and usable spectral range of the spectrograph were 1.2 nm and 380–760 nm, respectively. The wavelength and intensity of the spectrograph were calibrated with a mercury-argon lamp (Ocean Optics) and a halogen lamp with a known color-temperature (Thorlabs). We note that due to the low light intensity of the color-temperature lamp in the blue region of the spectrum and the short maximum integration time of the spectrograph (1 ms), the spectrum could not be corrected for instrument response below $\sim 460 \text{ nm}$. The spectrograph was triggered by the fire control circuits used to detonate the explosive charges. Spectra were recorded at an integration period of 15 μs per scan. Each shot was repeated several times in order to confirm reproducibility, and the light transmission of the fiber optic was checked after each shot. Damaged fibers were replaced prior to firing the next shot as necessary.

III. RESULTS AND DISCUSSION

In Figure 2(a), we show a typical data set for the horizontal fiber located at 90 cm. This fiber optic collects light from the outermost surface layers of the fireball during its evolution. Consistent with our earlier measurement,¹² for the first 15 μs following the start of detonation, little light is observed. Subsequent scans (later delay times) contain Al $^2P_{1/2} \leftarrow ^2S_{1/2}$ and $^2P_{3/2} \leftarrow ^2S_{1/2}$ atomic emissions at 394 and 396 nm, respectively,^{19,20} the AIO $X \leftarrow B$ emission band²¹ from 435 to 545 nm, and a large broadband emission. Observation of Al atomic emission peaks, the AIO vibronic band, and a broadband emission is typical of aluminum combustion.^{22–26} At later times, the Al and AIO emissions begin to fade away as the early-time free/gaseous Al combustion

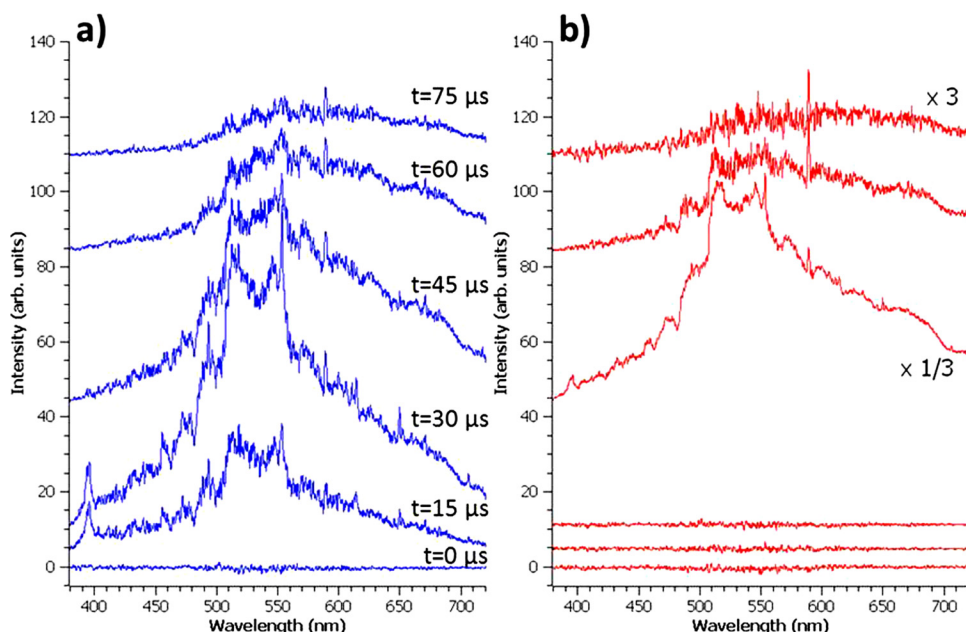


FIG. 2. Time-resolved emission spectra obtained from detonation of 20 g charges of RDX containing 20 wt. % aluminum nanoparticles. The charges were doped with 1 wt. % barium nitrate. Spectra in (a) were collected from the fiber optic located at 90 cm, facing the explosive charge. Delay times relative to the detonator signal are indicated. Spectra in (b) were obtained from the fiber optic at 10 cm, which faced upwards. Delays for each vertically shifted spectrum are the same as in (a). In both sets, a large broadband emission is evident. Superimposed upon the broadband are Al atomic emission lines found at 394 and 396 nm, an AIO vibronic band from 435 to 545 nm, and a Na atomic emission at 589 nm. Ba and Ba^+ lines are found at 455, 493, 554, 614, 650, and 706 nm. All spectra are plotted on the same intensity scale. Two of the spectra in (b) were rescaled as marked.

TABLE I. Apparent temperatures obtained from the Ba atomic emission lines. Estimated errors at the 95% confidence level are given in parentheses.

Time (μ s)	Fiber at 10 cm	Fiber at 90 cm
0
15	...	4000 (500)
30	...	3900 (400)
45	4000 (200)	...

processes halt, leaving only a broadband emission from hot particulates.¹² In several of the scans, we also see a Na emission at 589 nm resulting from Na impurities²⁷ in the sample, as well as peaks at 554 nm and 706 nm due to the $^1S_0 \leftarrow ^1P_1$ and $^3D_3 \leftarrow ^3F_4$ transitions in Ba atoms, and peaks at 455, 493, and 614 nm from the $^2S_{1/2} \leftarrow ^2P_{3/2}$, the $^2S_{1/2} \leftarrow ^2P_{1/2}$, and the $^2D_{5/2} \leftarrow ^2P_{3/2}$ transitions in Ba⁺ ions, respectively.²⁸ The peak at 650 nm may have contributions²⁸ from both Ba ($^3D_3 \leftarrow ^3D_3$) and Ba⁺ ($^2D_{3/2} \leftarrow ^2P_{1/2}$) at the resolution of the spectrograph.

In Figure 2(b), we see the signals recorded from the vertical fiber optic placed at 10 cm. In the first three scans, corresponding to times prior to arrival of the flame front at the fiber, no light is collected. In the scan beginning at 45 μ s, the flame front has arrived at the fiber and a large signal is observed. Light is also collected during subsequent scans, corresponding to luminescence from material in the interior of the fireball. It is interesting to compare these spectra with those collected from outside the fireball, shown in Figure 2(a). We see that once the flame front has arrived and the swept over the fiber, the spectra collected are remarkably similar to those in Figure 2(a), although the absolute intensities differ. The peaks and bands observed in the spectra in Figure 2(a), the underlying broadband emissions, and the general contours of the spectra are all reproduced in the spectra in Figure 2(b), albeit with a different signal-to-noise ratio. The apparent temperatures of the flame (calculated from the Ba atomic emission lines^{11,12}) are also similar and are shown in Table I. Regardless of which fiber optic is used to collect the spectrum, the apparent temperature of the spectrum is in the vicinity of 4000 K, in excellent agreement with earlier measurements.¹²

When the fiber is moved to 30 cm from the explosive charge, a similar behavior is observed. In Figure 3, we show the spectra collected from the vertical fiber at this position as well as the analogous spectra from the horizontal fiber at 90 cm. In Figure 3(a), we see the spectra from the fiber at 90 cm beginning at $t = 105 \mu$ s. By this time, free/gaseous Al combustion has ceased and the fireball has begun to cool;¹² the broadband emission and a Na line at ~ 589 nm are the only spectral features remaining. In subsequent spectra, these too fade in intensity. Unfortunately, the fireball is not sufficiently hot at this point to efficiently populate the upper Ba energy levels and permit a temperature measurement. Turning our attention to Figure 3(b), we find an empty spectrum at $t = 105 \mu$ s. Actually, a very small contribution may be present at ~ 550 nm, although it is difficult to be certain given the noise level in this region. All spectra collected from the fiber prior to this time are simply blank. In the spectra obtained at 120 μ s and later, we observe a broadband emission and the Na line as the flame front arrives and passes over the fiber. A comparison of the spectra in Figure 3(a) with those in Figure 3(b) reveals that once again the two sets of spectra are quite similar.

At first glance, the fact that the normalized spectra collected from inside and outside the fireball are consistently comparable to one another would seem to imply that the dynamics inside the fireball are also comparable to those near its surface. Hence, any spectra collected from outside might be regarded as representative of the overall dynamics. This impression is misleading, however, since it fails to account for the differences in the absolute spectral intensities. In Figure 2, we find that the ratios of the absolute intensities of the spectra in Figure 2(a) to those in Figure 2(b) are not constant. The spectrum at $t = 45 \mu$ s in Figure 2(b) is approximately a factor of 3 more intense than the corresponding spectrum in Figure 2(a). At $t = 60 \mu$ s, however, the spectrum in Figure 2(b) is of comparable intensity to that in Figure 2(a). And at 75 μ s, the spectrum in Figure 2(b) is now a factor of 3 weaker than that in Figure 2(a). Subsequent spectra become yet weaker. On the other hand, in Figure 3, we find that the absolute intensities of the spectra in Figure 3(a) and those in 3(b) (at least once the flame arrives at the fiber) are always

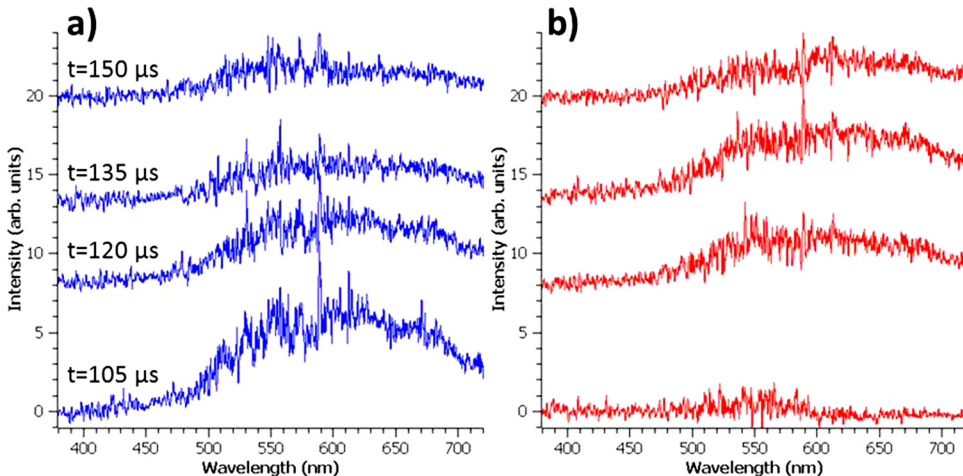


FIG. 3. Time-resolved emission spectra obtained from detonation of 20 g charges of RDX containing 20 wt. % aluminum nanoparticles and doped with 1 wt. % barium nitrate. Spectra in (a) were collected from the fiber optic located at 90 cm. Spectra in (b) were obtained from the fiber optic at 30 cm. Delay times are indicated relative to the detonator signal. All spectra are plotted on the same intensity scale.

comparable. This is in direct contrast to the trend we observed in Figure 2.

We know that following detonation, expansion of the fireball will change its composition as the detonation products mix with the surrounding air. At some point in its early evolution, the under-oxidized detonation products that constitute the fuel for the fireball will be primarily located behind the flame front. During such time, combustion would take place predominantly near the surface of the fireball as the hot fuel and air combine there, with little reaction possible in the interior due to lack of oxygen. If this is the case, we would expect the spectrum to be quite bright when the flame front arrives at the fiber and much less intense when the fiber is located behind the advancing flame front. This is precisely what we observe from the fiber located at 10 cm. At some point later in the evolution of the fireball, it will have expanded and mixed the hot fuels inside with surrounding air, and we would expect combustion to occur throughout. Hence, we would no longer expect the spectrum collected from a stationary point in the interior to rapidly weaken as the flame front advances. This is indeed what we observe from the fiber at 30 cm. Note that by the time the flame front reaches this fiber, the volume of the fireball has increased by a factor of ~ 27 compared to when it passed the fiber at 10 cm.

The fact that the normalized emission spectrum (and apparent temperatures) obtained from the fiber optics placed inside is virtually identical to those obtained from outside the fireball is also consistent with this hypothesis. In the early time evolution, they agree simply because the surface of the fireball is the only region undergoing combustion and light emission. In the late time, the fireball is well-mixed, and there should be little difference between the chemistry occurring in the interior and that near the surface. Thus, the respective emission spectra should again be consistent with one another.

IV. CONCLUSIONS

Taken together, the results of the present investigation suggest that while emission spectra collected from the surface of the fireball may be regarded as representative of the ongoing combustion processes in the fireball, they should not be taken as representative of the environment throughout until several volume expansions have occurred. Our data suggest that early time combustion processes (and light emission) are predominantly confined to the material near the surface, while the interior is dark. For the 20 g RDX/Al charges studied, this seems to be the case for $t \leq 60 \mu\text{s}$ and $r \leq 10 \text{ cm}$. Once the fuel in the fireball is well-mixed with the surrounding air, combustion appears to occur throughout, and we find no significant differences between the spectra collected from the interior of the fireball versus those from its surface. In the present study, this corresponds to $t \geq 120 \mu\text{s}$ or $r \geq 30 \text{ cm}$. In future work, it may be possible to utilize absorption spectroscopy to sample the apparently

non-combusting material behind the early-time flame front and to quantify the presumably cooler conditions there.

ACKNOWLEDGMENTS

The authors gratefully acknowledge funding and facilities provided for this work by the Air Force Research Laboratory under the NanoEnergetics Program. We would like to thank Mr. Rick Beesley and Mr. Mark Grimmonpre for their assistance in conducting these experiments.

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